Aging of residual surface resistance of superconducting lead cavities

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in Eq. (2) is negligible if $R\,dC/dt \ll 1$, or if $(d\epsilon/dt) \times [d(\Delta T)/dt] \ll L/AR$. Because $d(\Delta T)/dt \leq \Delta T_s/\tau_T$ and, according to Curie-Weiss law,[11] $d\epsilon/dT = \epsilon(|T - T_c|)^{-1}$, dC/dt can be neglected if

$$\theta \Delta T_s \gg T_c - T. \tag{8}$$

Thus for $\theta < 1$, condition (7) contains (8) and is sufficient for the validity of relations (5) and (5a).

Condition (7) means that one may approach T_c closer, the smaller ΔT_s becomes; e.g., if $\Delta T_s \approx 0.1\,°C$, $T_c - T$ may be ~1 °C. Because the rises of C_T and G_T with T are small (compared to those of λ and ϵ), τ_T remains constant in a range considerably closer to T_c. The measurements of λ and ϵ can thus be extended by using R_L small enough to maintain $\theta \ll 1$. Only during the rise of $V(t)$, for $\theta \ll 1$, does one have $\Delta T/\Delta T_s \approx \tau_e/\tau_T$—i.e., $\Delta T \approx \theta \Delta T_s$—and is condition (8) sufficient for the validity of relations (5) and (5a). For example, with $\Delta T_s = 0.1\,°C$ and $\theta = 0.01$, one can approach T_c to ~0.01 °C.

Measurements were carried out on six samples, 15—70 μm thick, of single-crystalline triglycine sulphate[12] (TGS) with major faces oriented perpendicular to the PE axis and provided with ir transparent electrodes. The samples were placed in a small oven with a CdS window. The oven was thermistor controlled with a temperature stability better than 0.05 °C. The temperature of the sample in the oven was measured within better than 0.1 °C with a calibrated thermocouple attached very close to the rear electrode. The whole mount was held in a vacuum-tight cryostat with a KBr window. The radiation source was a 500 °K blackbody with a shutter fast enough to provide step signals. The PE responses were measured on a Tektronix RM 564 storage cathode-ray oscilloscope (CRO) with a 3A9 amplifier of useable sensitivity at 50 μV/cm and with $R_L = 10^7\ \Omega$ while R_S was ~$10^{12}\ \Omega$.

The values of ϵ and λ, obtained from the PE response transients with the use of relations (5) and (5a) are shown in Fig. 4. The c curve in the figure is based on Refs. 9 and 10, and T_c is ~49.5 °C. The data of ϵ above T_c are obtained from the small residual PE responses.[13] Figure 5 is a plot of ϵ^{-1} versus T in the vicinity of T_c and shows that the values obtained for ϵ in samples T-18 and T-24 fit the Curie-Weiss law well and yield a Curie constant[11] of 3200 °K. A less typical value of ~5000 °K is found in sample Z-3.

The plot of λ^{-2} vs T in the vicinity of T_c is shown in Fig. 6. It fits the thermodynamic theory of a second-order ferroelectric transition[11,14] and has a slope of $8 \times 10^{12}\ (\mathrm{C/cm^2\,deg^{1/2}})^{-2}$. The agreement between our data and the previously reported experimental and theoretical results shows that the present method can be a useful tool in the study of ferroelectrics. A detailed discussion will be presented together with an analysis of the temperature dependence of the PE voltage response to step infrared signals in ferroelectrics.[15]

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[1] A. G. Chynoweth, Phys. Rev. 117, 1235 (1960); J. Appl. Phys. 27, 78 (1956).
[2] A. M. Glass, Phys. Rev. **172**, 564 (1968).
[3] A. Savage, J. Appl. Phys. 37, 3071 (1966).
[4] L. S. Kremenchugskii and V. B. Samoilov, Sov. Phys. Cryst. 12, 940 (1968); M. D. Kladkevich, L. S. Kremenchugskii, and A. F. Mal'nev, Ukr. Phys. J. 13, 442 (1968).
[5] G. Chanussot and Z. Malek, Compt. Rend. B 270, 844 (1970).
[6] N. D. Gavrilova, Yu. A. Zvirqzd, V. K. Novik, and V. G. Poshin, Sov. Phys. Solid State **13**, 1506 (1971).
[7] S. B. Lang, Phys. Rev. B 4, 3603 (1971); S. B. Lang and F. Steckel, Rev. Sci. Instr. **36**, 929 (1965).
[8] M. Simhony and A. Shaulov, J. Appl. Phys. 42, 3741 (1971).
[9] J. A. Gonzalo, Phys. Rev. Letters 21, 749 (1968); M. J. Tello and J. A. Gonzalo, J. Phys. Soc. Japan Suppl. **28**, 199 (1970).
[10] S. A. Taraskin, B. A. Strukov, and V. A. Meleshina, Sov. Phys. Solid State 12, 1089 (1970).
[11] F. Jona and G. Shirane, *Ferroelectric Crystals* (Pergamon, New York, 1962), pp. 31—34, 116.
[12] Laboratory grown; see V. Konstantinova, I. M. Silvestrova, and K. S. Aleksandrov, Sov. Phys. Cryst. 4, 63 (1960).
[13] Such responses were also observed by Chynoweth and by Glass (Ref. 2).
[14] E. Fatuzzo and W. J. Merz, *Ferroelectricity* (North-Holland, Amsterdam, 1967).
[15] M. Simhony, A. Shaulov, and A. Maman (unpublished); A. Hadni, Y. M. Henniger, R. Thomas, P. Vergnat, and B. Wyncke, Compt. Rend. 260, 4186 (1965).

Ageing of the residual surface resistance of superconducting lead cavities

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Measurements of the residual surface resistance of superconducting lead cavities as a function of time during a period of a month showed an oscillating variation. An explanation of the ageing curves is proposed.

The origin of the residual surface resistance of a superconducting surface has earlier been investigated experimentally by measurements of the dependence of the residual losses in superconducting lead and niobium cavities on frequency,[1-3] the mode in the cavity,[2-4] the applied magnetic field,[1,5-7] and the treatment of the surface.[2,4,7-10] Some authors have observed a change of the residual surface resistance with time.[2,9] Long time measurements on niobium cavities have been reported.[11] In the present work, long time measurements on lead

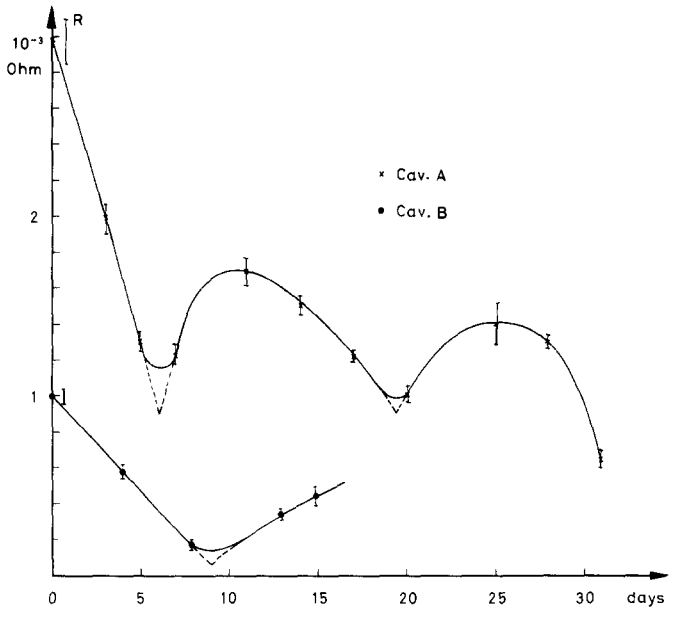

FIG. 1. Residual surface resistance of two superconducting cavities as a function of time after emersing them into gaseous helium.

cavities are reported, and a possible explanation of the results is presented.

The experiments in the present work were performed on two circular cylindrical cavities operating in the TE_{011} mode with the geometry factor 706 Ω, indicating the product of the surface resistance and the internal Q.

The cavities were constructed by machining a lead billet. The joints were placed at the corners, where the current is zero, in order to remove their influence.

The measurements were made at 35 GHz as reflection measurements. The residual surface resistance of the two cavities was determined as a function of time during periods of 30 and 15 days, respectively (Fig. 1). The measurement procedure was as follows: A curve of the surface resistance as a function of the temperature was taken on each of the days when measurements were performed. The curve was extrapolated to 0 °K in order to find the temperature-independent residual resistance. The measurements were repeated with intervals of two to five days. On the days when measurements were not carried out, the cavity was heated to room temperature. All the time, the cavity was in gaseous helium. The coupling to the cavity was left unchanged.

On each day of measurement, the cavity was evacuated to 10^{-3} Torr and refilled with gaseous helium while still at room temperature. After cooling to liquid-helium temperature, the cavity was evacuated again, following which the pressure was fixed at 0.05 Torr at 4.2 °K and the test vessel containing the cavity was closed. As a result the mass of the helium in the cavity was nearly independent of the temperature during the measurements.

The cavities had been stored in atmospheric air before the measurements started.

The following main characteristics of the measured curves can be seen from Fig. 1: (i) The minima have a tendency to be sharp; (ii) the slope at the beginning of the curves is negative; (iii) the maxima of the curves decrease with increasing time.

Heat treatment of one of the cavities A at 200 °C and 10^{-5} Torr removed the time variation almost totally and lowered the residual surface resistance to 1.4×10^{-4} Ω, corresponding to a cavity Q of 5×10^{6}, from which it has been concluded that the mentioned behavior of the residual resistance is due to contamination and work damage of the surface.

The loss mechanism leading to the measured curves has not been fully understood, but it is suggested that the variation stems from the contamination of the surface, especially the compounds formed by the gasses in the atmosphere and the lead.

The sharp minima can be explained by a mechanism where the contamination material in the cavity has a conductivity due to different charge carriers.[7] The ionization centers corresponding to carriers with opposite signs compensate each other as in a doped semiconductor where the ionization energy of the doping centers is much less than kT (≈0.1 meV at 1 °K) and/or the width of the energy doping level is broadened to such an extent that it overlaps the valence or conduction bands. This makes the conductivity slowly varying with temperature. Such a semiconductor model can explain the laps of the measured curves. The residual resistance is proportional to the conductivity, which in turn is proportional to the numerical value of the difference between the total density of ionized acceptor and donor centers, $|n_A - n_D|$. If n_A and n_D decay with different decay velocities, $|n_A - n_D|$ will reach a sharp zero. Several sharp minima such as those measured (Fig. 1) can occur if n_A and n_D are made up of a sum of several kinds of ionization centers, decaying with different velocities.

The slope at the origin has the tendency always to be negative in the "semiconductor" model if the buildup and decay processes of ionization centers are reversible. Those centers whose buildup velocity in atmospheric air is large will tend to have the largest density, which in turn will decay fastest, when the cavity is exposed to an inactive gas (or vacuum). This explains the negative slope at the origin of the measured curves.

The decrease of the maxima with time can be explained by the decrease of the total density of ionization centers with time.

The losses according to the proposed model are in agreement with previous measurements, showing that the residual losses are proportional to the frequency squared[1-3] and are larger in TM modes than in TE_{01m} modes.[2-4] This can easily be shown if the contamination layer is taken to be an Ohmic conducting layer backed up by an ideal conductor (superconductor). Both an electric field parallel to and an electric field perpendicular to the surface give a frequency-squared dependence of the residual losses. In addition, fissures in the surface parallel to the magnetic field, which are filled with Ohmic conducting material, also give residual losses proportional to the frequency squared. This is because

an electric field is created in the fissure, which is proportional to the frequency.[3]

Since the parameters involved in the contamination problem are very uncertain, estimations of the magnitude of losses must be crude. However, it can be seen for reasonable values of layer thickness and a conductivity $\sigma \approx 1-100\ (\Omega\,\mathrm{m})^{-1}$, which is representative for semiconductors, that the order of magnitude of the calculated residual losses agrees with those measured. In addition, for reasonable values of layer thickness (1 μ) and size (10 μ) of fissures in the surface, it can be shown that an electric field perpendicular to the surface can create losses which are of the same order of magnitude as, or one order of magnitude higher than, those coming from the fissuring. This is in agreement with measurements.[2]

However, the model has a number of unsolved questions. Thus, the structure and composition of the contamination material are unknown. Furthermore, it is not known how the proximity effect influences the microwave losses in the lossy material.

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[1]P. Kneisel, O. Stoltz, and J. Halbritter, IEEE Trans. Nucl. Sci. **NS-18**, 159 (1971).
[2]P. Flécher, J. Halbritter, R. Hietschold, P. Kneisel, W. Kühn, and O. Stoltz, IEEE Trans. Nucl. Sci. **NS-16**, 1018 (1969).
[3]J. Halbritter, J. Appl. Phys. 42, 82 (1971).
[4]J.P. Turneaure and I. Weissman, J. Appl. Phys. 39, 4417 (1968).
[5]J. Halbritter and K. Hoffman, Kernforschungzentrum Karlsruhe Report No. 3/67-9, 1967 (unpublished).
[6]J.M. Pierce, Ph.D. thesis (Stanford University, Stanford, Calif., 1967) (unpublished).
[7]M. Danielsen, Ph.D. thesis (The Technical University of Denmark, 1971) (unpublished).
[8]H. Hahn, H.J. Halama, and E.H. Foster, J. Appl. Phys. 39, 2606 (1968).
[9]H. Hahn and H.J. Halama, IEEE Trans. Nucl. Sci. **NS-16**, 1013 (1969).
[10]J.P. Turneaure and N. Tuong Viet, Appl. Phys. Letters 16, 333 (1970).
[11]H. Hahn and H.J. Halama, *Proceedings of the 7th International Conference on High Energy Accelerators in Yerevan* (Publishing House of the Academy of Science of the Armenian SSR, 1969), Vol. 2, p. 674.

High-efficiency $Ga_{1-x}Al_xAs$-GaAs solar cells

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Heterojunction solar cells consisting of $p\mathrm{Ga_{1-x}Al_xAs}$-pGaAs-nGaAs are grown by liquid-phase epitaxy and exhibit power conversion efficiencies of over 16% (corrected for contact area) measured in sunlight for air mass 1 at sea level, while efficiencies of 19—20% are obtained for an air mass value of 2 or more. The improved efficiencies compared to conventional homojunction (Si and GaAs) cells are attributed to the reduction of series resistance and the reduction of surface recombination losses resulting from the presence of the heavily doped $\mathrm{Ga_{1-x}Al_xAs}$ layer. Open-circuit voltages of 0.98—1.0 V and short-circuit currents of 18—21 mA/cm^2 (corrected for contact area) are observed for a solar input intensity of 98.3 mW/cm^2.

The $\mathrm{Ga_{1-x}Al_xAs}$-GaAs heterojunction system is probably unique among heterojunctions in that very few interface states are expected to exist at the boundary between the two materials because of their extremely close lattice match, while at the same time a growth technique is available that is capable of producing high-quality single-crystal material of controlled doping level. The use of a $\mathrm{Ga_{1-x}Al_xAs}$ layer on a GaAs substrate is therefore very attractive for solar-cell applications since the layer can be made thick and heavily doped to reduce the series resistance, will be transparent to most wavelengths which are absorbed efficiently by the GaAs, and should greatly reduce the recombination velocity at the GaAs "surface" (i.e., the interface between the two materials). It is the surface recombination velocity which is the greatest cause of low power conversion efficiencies in conventional GaAs solar cells.

The heterojunction cells reported here are grown by liquid-phase epitaxy in a vertical growth system described previously[1]; preliminary electrical measurements on similar cells have also been described.[2] An n-type GaAs wafer doped with Si to a level of $(2-5)\times10^{17}$ cm^{-3} is placed in a recess located in a carbon substrate holder. A melt consisting of Ga, Al, GaAs chunks, and Zn is placed in a melt chamber adjacent to the wafer; the temperature of the apparatus is then brought to 900 °C in an atmosphere of hydrogen and held constant for a period of 1 h to allow all parts of the system to equilibrate. Initially, the substrate is entirely enclosed by the high-purity carbon to prevent an appreciable loss of As from the surface during the equilibrating period. At the end of this period, the substrate is rotated into position below the melt, and the system is cooled from 900 to 890 °C at a rate of 0.1 °C/min. At 890 °C the sample is rotated away from the melt, and the near equality of the substrate thickness to the depth of the recess in the carbon holder causes the melt to be wiped off the substrate surface, which prevents spurious growth when the sample is cooled down to room temperature.

The Al content of the melt was varied to obtain compositions in the grown layer of $x = 0.3$, 0.5, and 0.7,